

Perdew *et al.* Reply: The AM05 condition [1], to fit a density functional to the jellium surface exchange-correlation (xc) energy, leads to accurate surface energies but is not a “possible explanation” [2] for accurate lattice constants of solids. In Table I, we combine the local spin density approximation (LSDA) for exchange with a modified PBEsol correlation, in which the gradient coefficient is chosen to fit the correct PBEsol [3] surface xc energy. The resulting lattice constants are typically shorter and less realistic than those of PBEsol or even LSDA. (See also Ref. [3].)

Kohn and Mattsson [4] proposed the Airy gas as a paradigm for density functional theory. Vitos *et al.* [5] constructed a generalized gradient approximation (GGA) for exchange by fitting the Airy gas conventional exchange energy density. In the absence of an Airy gas correlation energy density, the AM05 condition was used to construct a GGA for correlation. The Airy gas may contain much of the physics of solids (although a GGA cannot capture the conventional-gauge correction to the LSDA *exchange energy density* in the slowly varying limit [6]). Thus, while PBEsol is more correct for the exchange energy of a slowly varying density (and quite different from AM05; see Fig. S1 of Ref. [3]), AM05 xc resembles [3] PBEsol xc for those densities where the errors of GGA exchange and correlation can reliably cancel, including solids with reduced density gradient s everywhere less than about one [3]. For alkali and alkaline-earth metals and alkali halides, however, AM05 lattice constants can be closer [7] to the original Perdew-Burke-Ernzerhof (PBE) GGA [8] than to PBEsol.

While “GGAs for solids” such as AM05 and PBEsol are useful, our results [3,9] are more general. (1) No single GGA can describe with high accuracy the properties of both solids (surface energies and lattice constants) and molecules (total and atomization energies). The original PBE [8] is biased toward a correct description of molecules as explained in Ref. [3], while PBEsol is biased toward solids. (2) A meta-GGA form is more flexible and computationally not much slower than a GGA, making it the natural successor of LSDA and GGA in applications. The Tao-Perdew-Staroverov-Scuseria (TPSS) meta-GGA [10]

TABLE I. Lattice constants (\AA) in LSDA for exchange ($\mu = 0$) and correlation ($\beta = 0$) and in a GGA ($\mu = 0$, $\beta = 0.002$) which fits the PBEsol jellium surface xc energy, compared to PBEsol ($\mu = 0.1234$, $\beta = 0.046$) and exact values [3].

Solid	LSDA	$\mu = 0$, $\beta = 0.002$	PBEsol	Exact
Na	4.049	4.020	4.159	4.210
Si	5.410	5.415	5.442	5.423
LiCl	4.968	4.994	5.072	5.090
MgO	4.178	4.190	4.229	4.197
Cu	3.530	3.519	3.578	3.596
Pd	3.851	3.845	3.888	3.877

predicts accurate surface, atomization, and total energies. Its lattice constants are better than those of LSDA or PBE but not as good as those of PBEsol. An improved meta-GGA exchange must recover the second-order gradient expansion not just for slowly varying densities but “over a wide range of density gradients” [3] relevant to solids. (3) Like PBEsol, a meta-GGA should also improve upon the TPSS and PBE energy differences between molecules that differ only in equilibrium geometry [11].

Many GGA variants might give accurate lattice constants, but fitting to the jellium surface xc energy does not guarantee this. Restoring the gradient expansion for exchange might not be necessary for good lattice constants for a limited class of solids but is needed to construct more universal approximations [3].

This work was supported by NSF and MTA.

John P. Perdew,¹ Adrienn Ruzsinszky,¹ Gábor I. Csonka,² Oleg A. Vydrov,³ Gustavo E. Scuseria,⁴

Lucian A. Constantin,¹ Xiaolan Zhou,¹ and Kieron Burke⁵

¹Department of Physics

Tulane University

New Orleans, Louisiana 70118, USA

²Department of Chemistry

Budapest University of Technology and Economics

H-1521 Budapest, Hungary

³Department of Chemistry

Massachusetts Institute of Technology

Cambridge, Massachusetts 02139, USA

⁴Department of Chemistry

Rice University

Houston, Texas 77005, USA

⁵Department of Chemistry

University of California

Irvine, California 92697, USA

Received 23 September 2008; published 5 December 2008

DOI: 10.1103/PhysRevLett.101.239702

PACS numbers: 71.15.Mb, 31.15.E-, 71.45.Gm

- [1] R. Armiento and A. E. Mattsson, Phys. Rev. B **72**, 085108 (2005).
- [2] A. E. Mattsson, R. Armiento, and T. R. Mattsson, preceding Comment, Phys. Rev. Lett. **101**, 239701 (2008).
- [3] J. P. Perdew *et al.*, Phys. Rev. Lett. **100**, 136406 (2008); EPAPS PRLTAO-100-016814; erratum (to be published).
- [4] W. Kohn and A. E. Mattsson, Phys. Rev. Lett. **81**, 3487 (1998).
- [5] L. Vitos *et al.*, Phys. Rev. B **62**, 10046 (2000).
- [6] J. P. Perdew and Y. Wang, in *Mathematics Applied to Science*, edited by J. A. Goldstein *et al.* (Academic, New York, 1988); R. Armiento and A. E. Mattsson, Phys. Rev. B **66**, 165117 (2002).
- [7] G. I. Csonka *et al.* (to be published).
- [8] J. P. Perdew, K. Burke, and M. Ernzerhof, Phys. Rev. Lett. **77**, 3865 (1996); **78**, 1396(E) (1997).
- [9] J. P. Perdew *et al.*, Phys. Rev. Lett. **97**, 223002 (2006).
- [10] J. Tao *et al.*, Phys. Rev. Lett. **91**, 146401 (2003).
- [11] G. I. Csonka *et al.*, J. Chem. Theory Comput. **4**, 888 (2008).